

**TRANSMITTAL LETTER TO THE  
UNITED STATES  
DESIGNATED/ELECTED OFFICE  
(DO/EO/US) CONCERNING A FILING  
UNDER 35 U.S.C. 371**

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INTERNATIONAL APPLICATION NO.  
PCT/JP98/02500INTERNATIONAL FILING DATE  
June 5, 1998PRIORITY DATE CLAIMED  
November 7, 1997

## TITLE OF INVENTION

AQUEOUS SUSPENDED AGRICULTURAL CHEMICAL COMPOSITION

## APPLICANT(S) FOR DO/EO/US

Katsushi SHIO, Shoji SUZUKI and Naoki MATSUMOTO

**301 Rec'd PCT/PTO 23 DEC 1999**

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

**Items 11. to 16. below concern other document(s) or information included:**

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.  
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A small entity statement.
16. ☐ Other items or information:

U.S. APPLICATION NO. (if known, see 37 C.F.R. 1.5) <b>09/446730</b>		INTERNATIONAL APPLICATION NO. PCT/JP98/02500		ATTORNEY'S DOCKET NUMBER 105081	
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17. <input checked="" type="checkbox"/> The following fees are submitted:  <b>Basic National fee (37 CFR 1.492(a)(1)-(5)):</b>  Search Report has been prepared by the EPO or JPO.....\$840.00  International preliminary examination fee paid to USPTO (37 CFR 1.482).....\$670.00  No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....\$690.00  Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO.....\$970.00  International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4).....\$ 96.00  <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				CALCULATIONS		PTO USE ONLY	
				\$840.00			
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$			
Claims	Number Filed	Number Extra	Rate				
Total Claims	4- 20 =	0	X \$ 18.00	\$			
Independent Claims	1- 3 =	0	X \$ 78.00	\$			
Multiple dependent claim(s)(if applicable)			+ \$260.00	\$			
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$840.00			
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				\$			
<b>SUBTOTAL =</b>				\$840.00			
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 month from the earliest claimed priority date (37 CFR 1.492(f)).				\$			
<b>TOTAL NATIONAL FEE =</b>				\$840.00			
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a. ☒ Check No. 105256 in the amount of \$840.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \$\_\_\_\_\_ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment, to Deposit Account No. 15-0461. A duplicate copy of this sheet is enclosed.

**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.**

SEND ALL CORRESPONDENCE TO:  
 OLIFF & BERRIDGE, PLC  
 P.O. Box 19928  
 Alexandria, Virginia 22320

  
 NAME: James A. Oliff  
 REGISTRATION NUMBER: 27,075  
  
 NAME: Thomas J. Pardini  
 REGISTRATION NUMBER: 30,411

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Katsushi SHOI, Shoji SUZUKI and Naoki MATSUMOTO

U.S. National Stage of PCT/JP98/02500

Filed: December 23, 1999

Docket No.: 105081

For: AQUEOUS SUSPENDED AGRICULTURAL CHEMICAL COMPOSITION

PRELIMINARY AMENDMENT

Assistant Commissioner of Patents  
Washington, D. C. 20231

Sir:

Prior to initial examination, please amend the above-identified application as follows:

IN THE CLAIMS:

Please amend the claims as follows:

Claim 4, line 2, delete "to 3".

REMARKS

Claims 1-4 are pending. By this Preliminary Amendment claim 4 is amended to eliminate multiple dependencies. No new matter is added. Prompt and favorable examination on the merits.

Respectfully submitted



James A. Oliff  
Registration No. 27,075

Thomas J. Pardini  
Registration No. 30,411

WPB/epb  
**OLIFF & BERRIDGE, PLC**  
**P.O. Box 19928**  
**Alexandria, Virginia 22320**  
**Telephone: (703) 836-6400**

## AQUEOUS SUSPENDED AGRICULTURAL CHEMICAL COMPOSITION

BACKGROUND OF THE INVENTION

## 1. Field of the Invention

The present invention relates to an aqueous suspended agricultural chemical composition having high preservation stability, which contains ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy ) phenoxy] propionate.

## 2. Description of the Related Art

Ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy ) phenoxy] propionate (hereinafter referred to as quizalofop-p-ethyl ) is known to have two kinds of crystal forms. One is a low melting point type crystal (hereinafter referred to as  $\alpha$ -type crystal) and the other is a high melting point type crystal (hereinafter referred to as  $\beta$ -type crystal)(see Japanese Patent Examined Publication No. Hei 4-76721).

Although quizalofop-p-ethyl has heretofore been handled as an emulsion, there is a demand for a highly safe aqueous suspended composition free of organic solvents (floable agent). However when the aqueous suspended composition having been prepared with industrially produced  $\alpha$ -type crystal quizalofop-p-ethyl particles is stored under severe condition, at 50 °C for 30 days, after the preparation, a phenomenon wherein flowability of this aqueous

suspended composition deteriorates during the storage is sometimes observed. Therefore there has been a demand for the further improvement of the preservation stability the aqueous suspended composition.

#### SUMMARY OF THE INVENTION

As a result of an intensive study by the present inventors in order to solve the above-mentioned problem, there is found that an aqueous suspended agricultural chemical composition containing quizalofop-p-ethyl wherein the ratio of the  $\beta$ -type crystal is at least 80 wt% or more, a surfactant, and water, had extremely good preservation stability. Thus, the present invention is completed. That is, the present invention relates to [1] to [4] enumerated hereunder.

[1] An aqueous suspended agricultural chemical composition containing the following components (a), (b), and (c).

(a) Ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy) phenoxy] propionate wherein the ratio of the  $\beta$ -type crystal is 80% by weight or more,

(b) Surfactant,

(c) Water.

[2] An aqueous suspended agricultural chemical composition described in the above item [1], wherein the component (a)

is ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy ) phenoxy] propionate wherein the ratio of  $\beta$ -type crystal is 85% by weight or more.

[3] An aqueous suspended agricultural chemical composition as described in the above item [1], wherein the component (a) is Ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy ) phenoxy] propionate wherein the ratio of  $\beta$ -type crystal is 90% by weight or more.

[4] An aqueous suspended agricultural chemical composition as described in the above item [1] to [3], wherein the component (a) is 1 to 60 parts by weight, the component (b) is 0.1 to 60 parts by weight, and the component (c) is 20 to 95 parts by weight, respectively, based on 100 parts by weight of the aqueous suspended composition.

In quizalofop-p-ethyl used in the aqueous suspended agricultural composition of the present invention, the ratio of  $\beta$ -type crystal is at least 80% by weight or more, preferably 85% by weight or more, and more preferably 90% by weight or more in terms of the preservation stability of the agricultural chemical composition. If the ratio of  $\beta$ -type crystal is less than 80wt%, the particle size of quizalofop-p-ethyl particle in the aqueous suspended agricultural chemical composition may increase (hereinafter

referred to as particle growth) or flowability of the composition may deteriorate by storing the composition under severe condition such as at 50°C for 30 days. The particle growth is not preferred in terms of herbicide effect and deteriorated flowability is not preferred in terms of the convenience of handling.

Quizalofop-p-ethyl is known to have two kinds of crystal forms. One is  $\alpha$ -type crystal (low melting point type crystal), a plate crystal, of which endothermal peak is around 74 °C in differential scanning calorimetry, and the other is  $\beta$ -type crystal (high melting point type crystal), needle crystal, which endothermal peak is around 80 °C .

These two crystal forms of quizalofop-p-ethyl are more easily discriminated each other through X-ray powder diffraction, differential scanning calorimetry, and microscopic observations.

Hereinafter, description is made of the characteristics of these two crystal forms.

	Melting point	Crystalline form	Characteristic X-ray diffraction peak
$\alpha$ -type crystal	74 to 76 °C	Plate crystal	$2\theta = 4.36, 8.68$
$\beta$ -type crystal	80 to 82 °C	Fine needle crystal	$2\theta = 5.32, 6.38$

As the weight ratio of  $\alpha$  -type crystal to  $\beta$  -type crystal approximates to the area ratio of each endothermal peak in the differential scanning calorimetry, the weight ratio of  $\alpha$  -type crystal to  $\beta$  -type crystal is obtained from the area ratio of the respective endothermal peaks.

The method of obtaining quizalofop-p-ethyl, in which the ratio of  $\beta$  -crystal is 80% by weight or more, is not specifically limited. For example, a method described in Japanese Patent Examined Publication No. Hei 4-76721 is suitably used in this invention. According to this method, quizalofop-p-ethyl containing  $\beta$  -type crystal at any ratio can be obtained.

Alternatively, quizalofop-p-ethyl may be replaced with diphenyl ether herbicide such as propaquizafop (general name), quizalofop-p-tefuryl (general name), fenoxaprop-ethyl (general name) in the invention. In this case,  $\beta$  -type



crystal means high melting point type crystal and  $\alpha$  -type crystal means low melting point type crystal.

Usable surfactant is not specifically limited and various anionic surfactants and nonionic surfactants heretofore used in agricultural chemical field are suitably used in the invention. Examples of surfactants are listed hereinafter. However surfactants suitably used in the invention are not limited to those listed therein.

Examples of anionic surfactants which can be suitably used in the invention include sulfonic acid surfactants, sulfate surfactants, phosphate surfactants and their salts. Examples of suitable sulfonic acid surfactants are alkylsulfonic acid, alkylolefinsulfonic acid, lignosulfonic acid, alkylbenzenesulfonic acid, alkyl naphthalenesulfonic acid, naphthalenesulfonic acid formaldehyde condensate, and dialkylsulfosuccinate. Examples of suitable sulfate surfactants are polyoxyethylene alkylether sulfate, polyoxyethylene alkylallylether sulfate, polyoxyethylene styrylphenylether sulfate, polyoxyethylene phenylalkylallylether sulfate, polyoxyalkylene glycol sulfate, higher alcohol sulfate, and fatty acid ester sulfate. Examples of suitable phosphate surfactants are polyoxyethylene alkylether phosphate, polyoxyethylene

alkylallyl phosphate, polyoxyethylene phenylalkylallylether phosphate, higher alcohol phosphate, and polyoxyethylene tribenzylphenol phosphate. Example of their salts are sodium-, potassium-, magnesium-, calcium-, ammonium-, ethanolamine-, diethanolamine- and triethanol amine-salt.

Examples of suitable nonionic surfactants are polyoxyethylene alkylallylether, polyoxyethylene styrylphenylether, polyoxyethylene alkylether, polyoxyethylene phenylalkylallylether, polyoxyethylene sorbitan fatty acid ester, polyoxyethylene glycol, polyoxyethylene polyoxypropylene block co-polymer, polyoxyalkylene glycol, alkyne diol (acetylene glycol), alkynylene polyoxyethylenediol, sorbitan fatty acid ester and alkylallylether formaldehyde condensate and so forth.

These surfactants can be used separately or two kinds or more surfactants are allowed to be mixed. The mixing ratio can be freely selected.

In the aqueous suspended agricultural chemical composition of the present invention, content of quizalofop-p-ethyl in which the ratio of  $\beta$ -type crystal is 80% by weight or more, is not specifically limited, however it is generally 1 to 60 parts by weight, preferably 3 to 50 parts by weight to 100 parts by weight of the composition.

Content of the surfactant is generally 0.1 to 60 parts by weight, preferably 0.3 to 50 parts by weight. Content of water is generally 20 to 95 parts by weight.

The composition of the invention can further contain following active components of agricultural chemicals.

Examples of the active components of agricultural chemicals suitably used in the invention include Diflufenican (general name), Propanil (general name), Dicamba (general name), Picloram (general name), 2,4-D (general name), 2,4-DB (general name), 2,4-DP (general name), Fluroxypyr (general name), MCPA (general name), MCPP (general name), Triclopyr (general name), Diclofop-methyl (general name), Fenoxaprop-ethyl (general name), Fluazifop-butyl (general name), Haloxyfop-methyl (general name), Chloridazon (general name), Norflurazon (general name), Chlorpropham (general name), Desmedipham (general name), Phenmedipham (general name), Propham (general name), Alachlor (general name), Acetochlor (general name), Butachlor (general name), Metazachlor (general name), Metolachlor (general name), Pretilachlor (general name), Propachlor (general name), Oryzalin (general name), Pendimethalin (general name), Trifluralin (general name), Acifluorfen (general name), Bifenox (general name), Fluoroglycofen (general name),

Fomesafen (general name), Halosafen (general name), Lactofen (general name), Oxyfluorfen (general name), Chlortoluron (general name), Diuron (general name), Fluometuron (general name), Isoproturon (general name), Linuron (general name), Metabenzthiazuron (general name), Lenacil (general name), Bromacil (general name), Imazapyr (general name), Imazaquin (general name), Imazethapyr (general name), Imazamethabenz (general name), Imazamox (general name), Alloxydim (general name), Clethodim (general name), Cycloxydim (general name), Sethoxydim (general name), Talkoxydim (general name), Bromoxynil (general name), Dichlobenil (general name), Ioxnil (general name), Mefenacet (general name), Amidosulfuron (general name), Bensulfuron-methyl (general name), Chlorimuron-ethyl (general name), Chlorsulfuron (general name), Cinosulfuron (general name), Metsulfuron-methyl (general name), Nicosulfuron (general name), Primisulfuron (general name), Prosulfuron (general name), Halosulfuron-methyl (general name), Thifensulfuron-methyl (general name), Triasulfuron (general name), Tribenuron-methyl (general name), Butylate (general name), Cycloate (general name), Diallylate (general name), EPTC (general name), Esprocarb (general name), Molinate (general name), Prosulfocarb (general name), Thiobencarb (general

name), Triallate (general name), Atrazine (general name),  
Cyanazine (general name), Simazine (general name), Simetryne  
(general name), Terbutryn (general name), Terbutylazin  
(general name), Hexazinon (general name), Metamitron (general  
name), Metribuzin (general name), Aminotriazole (general  
name), Benfuresate (general name), Bentazon (general name),  
Cinmethylin (general name), Clomazone (general name),  
Clopyralid (general name), Difenzoquat (general name),  
Dithiopyl (general name), Ethofumasate (general name),  
Fluorochloridone (general name), Glufosinate (general name),  
Glyphosate (general name), Isoxaben (general name), Paraquat  
(general name), Pyridate (general name), Quinclorac (general  
name), Quinmerac (general name), Sulphosate (general name),  
Tridiphane (general name), Flumetsulam (general name),  
Fluthiacet-methyl (general name), Sulfentrazone (general  
name), Carfentrazone (general name), Dimethenamide (general  
name), Isoxaflutole (general name), Oxasulfuron (general  
name), Cloransulam-methyl (general name), Flumiclorac-pentyl  
(general name), Fluthiamide (general name), Aclonifen  
(general name), Benazolin (general name).

These active components of agricultural chemicals are  
used separately or two kinds or more of them are allowed to  
be mixed. The mixing rate can be freely selected. The amount

of addition of the active components to the composition of the invention is properly selected. However, preferably, it is selected within 0.1 to 50 parts by weight.

Adjuvants suitably contained in the composition of this invention include thickeners, antifreezing agents, antifoaming agents, antibacterial and antifungal agents, and colorants, and the examples are cited as follows.

Thickeners suitably used in the composition of this invention are not specifically limited and include organic- and inorganic-natural materials and synthetic- and semisynthetic products. Examples of the thickeners suitably used in the invention include heteropolysaccharides, water soluble polymer compounds, cellulose derivatives, and smectite clay minerals. Examples of suitable heteropolysaccharides are xanthan gum, welan gum and rhamnan gum. Examples of suitable water soluble polymer compounds are polyvinylalcohol, polyvinylpyrrolidone, polyacrylic acid, sodium polyacrylate, and polyacrylamide. Examples of suitable cellulose derivatives are methylcellulose, carboxymethylcellulose, carboxyethylcellulose, hydroxyethylcellulose, and hydroxypropylcellulose. Examples of suitable smectite clay minerals are montmorillonite, saponite, hectorite, bentonite,

raponite and synthetic smectite. These thickeners can be used separately or two kinds or more of them can be mixed and the ratio of the mixing can be selected freely.

Intact thickeners or those dispersed in water beforehand are allowed to be added. And the amount of the addition can be selected freely at its addition to the component of the invention.

Antifreezing agents may be used in the composition of this invention if required. As usable antifreezing agents, there may be added ethylene glycol, diethylene glycol, propylene glycol, or the like. The amount of the addition is freely selected at its addition to the composition of this invention.

Furthermore, antifoaming agent such as silicon emulsion, antibacterial and antifungal agents, and colorants are allowed to be blended. Various antibacterial and antifungal agents may be used in the invention. Example of suitable antibacterial and antifungal agents include benzoic acid and its salt, Proxel GXL (ICI Corp.) and Proxel XL-2 (ICI Corp.). Other than that, proper selection is made from various antibacterial and antifungal agents. And the amounts of addition can be freely selected at their addition to the composition of the invention.

The composition of the invention is, for example, prepared by a method in which the solid components contained in the composition of the invention is mixed into the water added and mixed with a surfactant, and then the mixture is subjected to atomization with a wet mill such as a sand grinder, and then, to the atomized mixture, other adjuvants such as a thickener is added and mixed.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

Next, examples of the preparation of the composition of this invention and comparative examples of the aqueous mixtures of agricultural chemical compositions used for comparison are explained. "Part" used in examples and comparative examples means part by weight. Furthermore, this invention is not limited to these examples.

##### Example 1

##### 1. Preparation of ground slurry

4.5 parts of Sorpol 3353 (mixture of polyoxyethylene-styrylphenylether and polyoxyethylene-polyoxypropylene block copolymer/Toho Chemical Industry, Product name), 0.15 parts of Antifoam E-20 (Silicone antifoaming agent/Kao Corp., Product name) were dissolved in 59.35 parts of water. 36



parts of quizalofop-p-ethyl (ratio of  $\beta$  -type crystal: 100% by weight) was dispersed therein. Then the dispersion solution was subjected to wet milling with a sand-grinder ( AIMEX CO., Ltd.)

using 300g of glass beads (1.0 to 1.5 mm in diameter) for 120 minutes at 2,000 rpm to obtain ground slurry. During the milling, temperature of cooling water was kept within 10 to 15°C , and temperature of the solution was kept within 10 to 20 °C .

## 2..Preparation of dispersion medium

2.25 parts of Vangel (smectite clay mineral/thickener/R.T. VANDERBILT Corp., Product name), 0.14 parts of xanthan gum, and 0.14 parts of Proxycel GXL (ICI Corp., Product name) were dispersed in 67.48 parts of water in order of Vangel, xanthan gum and Proxycel GXL. Then 30 parts of propylene glycol is added therein to obtain a dispersion medium.

## 3. Preparation of an aqueous suspended agricultural chemical composition

The above-mentioned ground slurry was mixed in the dispersion medium in the ratio of two to one to obtain a homogeneous aqueous suspended agricultural chemical composition.

## 4. Confirmation of the crystalline form of .

quizalofop-p-ethyl in the aqueous suspended composition

From the obtained aqueous suspended agricultural chemical composition, about 3g of the composition was separated, and then it was suspended in about 60ml of water. Then the suspension was subjected to a centrifugal separator (Kokusan Enshinki Corp., H-300 type, at 3,000 rpm for 15 minutes) and the formed supernatant was removed. Then the obtained precipitation was suspended in about 60ml of water and subjected to the centrifugal separator again. This washing operation with water was repeated six times. The obtained precipitation was spread on a filter paper and then dried in a desiccator for about 20 hours. Then 3 to 5 mg of the dried material was weighed accurately and subjected to differential scanning calorimetry (Mac Science Corp. Differential scanning calorimeter 3100, heating rate = 1°C per minute, sampling every 0.3 second). From the area ratio of endothermal peak of  $\alpha$  - type crystal to that of  $\beta$  -type crystal, ratio of  $\beta$  -type crystal in quizalofop-p-ethyl was found to be 100% by weight.

#### Examples 2 to 5

According to the process of Example 1, aqueous suspensions of agricultural chemical composition, in which

the ratio of  $\beta$  -type crystal in quizalofop-p-ethyl is 80% by weight or more, were obtained (Example 2: 96wt%, Example 3: 90wt%, Example 4: 85wt%, Example 5: 80wt%).

#### Example 6

Example 1 was repeated using quizalofop-p-ethyl (ratio of  $\beta$  -type crystal: 100% by weight) except that 4.5 parts of Soprophor FL (polyoxyethylene tristyrylphenylether phosphate /RHONE-POULENC Corp., Product name) was used in stead of 4.5 parts of Sorpol 3353 to prepare an aqueous suspended agricultural chemical composition. Ratio of  $\beta$  -type crystal in quizalofop-p-ethyl was 100% by weight in the obtained aqueous suspended agricultural chemical composition.

#### Example 7

##### 1. Preparation of ground slurry

6.25 parts of propylene glycol, 3.75 parts of Sorpol 3353, and 0.13 parts of Nopco 8034L (Silicon antifoaming agent/ San Nopco Limited, Product name) were dissolved in 43.62 parts of water. In this solution, 46.25 parts of quizalofop-p-ethyl (ratio of  $\beta$  -type crystal: 100% by weight) was dispersed. Then the disperse solution was

subjected to wet milling by a sand grinder using 300g of glass beads (1.0 to 1.5mm in diameter) at 2,000 rpm, for 120 minutes to obtain ground slurry. During the milling, temperature of cooling water was kept within 5 to 10°C , and the temperature of the solution was kept within 10 to 20 °C .

## 2. Preparation of a dispersion medium

0.25 parts of rhamzan gum and 0.25 parts of Proxycel XL-2 (ICI Corp., Product name) were dispersed in 99.5 parts of water in order of rhamzan gum and Proxycel XL-2, to obtain a dispersion medium.

## 3. Preparation of an aqueous suspended agricultural chemical composition

The above-mentioned ground slurry and dispersion medium were mixed in the ratio of 4 to 1 to obtain a homogeneous aqueous suspended agricultural chemical composition. The rate of  $\beta$  -type crystal in quizalofop-p-ethyl was 100% by weight in the obtained aqueous suspended agricultural chemical composition.

### Example 8

According to the process of Example 7, an aqueous suspended agricultural chemical composition was obtained, in which a ratio of  $\beta$ -type crystal of quizalofop-p-ethyl was 96% by weight.

#### Example 9

##### 1. Preparation of ground slurry

5.56 parts of propylene glycol, 3.33 parts of Sorpol 3353, and 0.11 parts of Nopco 8034L were dissolved in 35.44 parts of water. 55.56 parts of quizalofop-p-ethyl (ratio of  $\beta$ -type crystal: 100% by weight) was dispersed in the solution. Dispersion solution was subjected to wet milling by a sand grinder using 300g of glass beads (1.0 to 1.5mm in diameter) at 2,000 rpm for 120 minutes to obtain ground slurry. During the milling, temperature of cooling water was kept within 5 to 10 °C and temperature of the solution was kept within 10 to 20 °C .

##### 2. Preparation of a dispersion medium

0.25 parts of welan gum and 0.25 parts of Proxel GXL were dispersed in order of Welan gum and Proxycel GXL, to obtain a dispersion medium.

### 3. Preparation of an aqueous suspended agricultural chemical composition

The above-mentioned ground slurry and dispersion medium was mixed in the ratio of 9 to 1 to obtain a homogeneous aqueous suspended agricultural chemical composition. Ratio of  $\beta$  - type crystal of quizalofop-p-ethyl is 100% by weight in the obtained aqueous suspended agricultural chemical composition.

#### Comparative Examples 1 to 4

According to the process of Example 1, an aqueous mixture of agricultural chemical composition was obtained, in which ratio of  $\beta$  -type crystal in quizalofop-p-ethyl was less than 80% by weight (Comparative Example 1: 78wt%, Comparative Example 2: 38wt%, Comparative Example 3: 10wt%, Comparative Example 4: 0wt%).

#### Test Example

Particle size and viscosity of the aqueous suspended agricultural chemical composition obtained in Example 1 to 9, and in Comparative Example 1 to 4 were measured. Further, after they had been put in vials (inside volume 50 ml) respectively and stored in thermostatic chamber at 50 °C

for 30 days, the particle size were measured.

## 1. Measurement of the particle size

Volume medium diameter (d50) values (  $\mu$  m) of the particles were measured with a Laser Diffraction Technique Particle Size Analyzer LS-130 (Coulter Corp.) using garnet. omd as an optical model.

## 2. Measurement of viscosity

The viscosity was measured with a Viscometer DV-III (Brookfield Corp.) using No.2 rotor at 30 rpm and 25°C .

Table 1 shows the result. The symbols in the Table have the following meanings.

A: A ratio (wt%) of  $\beta$  -type crystal of quizalofop-p-ethyl in the aqueous suspended composition.

B: Volume medium diameter( $\mu$  -m) of quizalofop-p-ethyl particle in the aqueous suspended composition immediately after preparation.

C: Viscosity (cps) of the aqueous suspended composition immediately after preparation.

D: Volume medium diameter( $\mu$  -m) of the particle in the aqueous suspended composition after 30 days storage at 50°C.

E: Viscosity (cps) of the aqueous suspended composition after 30 days storage at 50°C . .

Table 1

	A	B	C	D	E
Example 1	100	1.3	180	1.9	248
Example 2	96	1.4	213	1.8	321
Example 3	90	1.2	167	2.0	252
Example 4	85	1.3	216	2.2	279
Example 5	80	1.3	193	2.4	332
Example 6	100	1.1	178	1.6	232
Example 7	100	1.0	202	1.7	185
Example 8	96	1.2	227	1.6	178
Example 9	100	1.3	451	2.1	337
Comparison 1	78	1.2	178	4.3	>1000
Comparison 2	38	1.4	201	5.8	>1000
Comparison 3	10	1.1	194	4.9	>1000
Comparison 4	0	1.2	225	5.2	>1000

B and C show physical properties immediately after preparation.

D and E show physical properties after 30 days storage at 50°C .

Above Table shows that, if the ratio of  $\beta$  -type crystal of quizalofop-p-ethyl is 80% by weight or more, the



preservation stability of the aqueous suspended composition was high.

Because the Table shows that, even after severe storage condition, 50 °C for 30 days, the flowability was kept well making the composition convenient to handle and the growth of quizalofop-p-ethyl particle was small making the herbicidal activity high.

#### Effect of the Invention

An aqueous suspended agriculture chemical composition in the invention maintains good flowability and the growth of the quizalofop-p-ethyl particle is small, resulting in high preservation stability of the compositions under severe preservation condition.

What is claimed is:

1. An aqueous suspended agricultural chemical composition containing following components (a), (b) and (c),

(a) Ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy)phenoxy] propionate wherein the ratio of  $\beta$  -type crystal is 80% by weight or more,

(b) Surfactant,

(c) Water.

2. An aqueous suspended agricultural chemical composition according to claim 1, wherein the component (a) is ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy)phenoxy] propionate of which the ratio of  $\beta$  -type crystal is 85% by weight or more.

3. An aqueous suspended agricultural chemical composition according to claim 1, wherein the component (a) is ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy)phenoxy] propionate of which the ratio of  $\beta$  -type crystal is 90% by weight or more.

4. An aqueous suspended agricultural chemical composition according to claim 1 to 3, wherein the component (a) is 1 to 60 parts by weight, the component (b) is 0.1 to 60 parts by weight, and the component (c) is 20 to 95 parts by weight, respectively, based on 100 parts of aqueous suspended composition.

# ABSTRACT OF THE DISCLOSURE

The present invention relates to an aqueous suspended agricultural chemical composition wherein component (a) is ethyl (R)-2-[4-(6-chloroquinoxalin-2-yloxy) phenoxy] propionate of which the ratio of  $\beta$  -type crystal is 80% by weight or more, component (b) is surfactant and component (c) is water. An aqueous suspended agricultural chemical composition of the invention has good flowability and the growth of quizalofop-p-ethyl particle is small, resulting in high preservation stability under severe preservation condition.

**DECLARATION AND POWER OF ATTORNEY  
UNDER 35 USC § 371(c)(4) FOR  
PCT APPLICATION FOR UNITED STATES PATENT**

As a below named inventor, I hereby declare that:  
my residence, post office address and citizenship are as stated below under my name:

I verily believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought, namely the invention entitled:

AQUEOUS SUSPENDED AGRICULTURAL CHEMICAL COMPOSITION

described and claimed in international application number PCT/JP98/02500 filed on JUNE 5, 1998

I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations §1.56.

Under Title 35, U.S. Code §119, the priority benefits of the following foreign application(s) filed within one year prior to my international application are hereby claimed:

Japanese Patent Application No. Hei 9-186558 filed on July 11, 1997

Japanese Patent Application No. Hei 10-118457 filed on April 28, 1998

The following application(s) for patent or inventor's certificate on this invention were filed in countries foreign to the United States of America either (a) more than one year prior to my international application, or (b) before the filing date of the above-named foreign priority application(s)

I hereby appoint the following as my attorneys of record with full power of substitution and revocation to prosecute this application and to transact all business in the Patent Office:

James A. Oliff, Reg.No.27,075; William P. Berridge, Reg.No.30,024;  
Kirk M. Hudson, Reg.No.27,562; Thomas J. Pardini, Reg.No.30,411;  
Edward P. Walker, Reg.No.31,450; Robert A. Miller, Reg.No.32,771;  
Mario A. Costantino, Reg.No.33,565; and Caroline D. Dennison, Reg.No.34,494.

**ALL CORRESPONDENCE IN CONNECTION WITH THIS APPLICATION SHOULD BE SENT TO OLIFF & BERRIDGE PLC, P.O. BOX 19928, ALEXANDRIA, VIRGINIA, 22320, TELEPHONE (703) 836-6400.**

I hereby declare that I have reviewed and understand the contents of this Declaration, and that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Typewritten Full Name

of Sole or First Inventor	<u>Katsushi</u>	<u>SHIO</u>
Given Name	<u>Katsushi Shio</u>	Family Name
**Inventor's Signature	<u>December</u>	<u>27</u>
** Date of Signature	<u>Month</u>	<u>1999</u>
	<u>Day</u>	<u>Year</u>
Residence	<u>Funabashi-shi</u>	<u>Chiba-ken</u>
City		State of Province
Citizenship	<u>Japanese</u>	<u>JPV</u> Country
Post Office Address	<u>c/o Nissan Chemical Industries, Ltd. Central Research Institute, 722-1, Teuboi-cho,</u>	
(Insert complete mailing address, including country)	<u>Funabashi-shi, Chiba-ken 274-8507, Japan</u>	

\*Note to Inventor: Please sign name on line 2 exactly as it appears line 1 and insert the actual date of signing on line 3.

IF THERE IS MORE THAN ONE INVENTOR USE PAGE 2 AND PLACE AN "X" HERE ☒

(Discard this page in a sole inventor application)

2-6 ✓  
Typewritten Full Name  
of Second Joint

Inventor (if any) Shoji SUZUKI  
Given Name Middle Initial Family Name

\*\*Inventor's Signature Shoji Suzuki  
\*Date of Signature December 27 1999  
Month Day Year

Residence Funabashi-shi Chiba-ken Japan  
City State or Province Country

Citizenship Japanese JPV

Post Office Address c/o Nissan Chemical Industries, Ltd. Central Research Institute, 722-1, Tsuboi-cho.

(Insert completing mailing Funabashi-shi, Chiba-ken 274-8507, Japan  
address, including country)

3-0 ✓  
Typewritten Full Name  
of Third Joint

Inventor (if any) Naoki MATSUMOTO  
Given Name Middle Initial Family Name

\*\*Inventor's Signature Naoki Matsumoto  
\*Date of Signature December 27 1999  
Month Day Year

Residence Funabashi-shi Chiba-ken Japan  
City State or Province Country

Citizenship Japanese JPX

Post Office Address c/o Nissan Chemical Industries, Ltd. Central Research Institute, 722-1, Tsuboi-cho.

(Insert completing mailing Funabashi-shi, Chiba-ken 274-8507, Japan  
address, including country)

Typewritten Full Name  
of Fourth Joint

Inventor (if any) \_\_\_\_\_  
Given Name Middle Initial Family Name

\*\*Inventor's Signature \_\_\_\_\_  
\*Date of Signature \_\_\_\_\_  
Month Day Year

Residence \_\_\_\_\_  
City State or Province Country

Citizenship \_\_\_\_\_

Post Office Address \_\_\_\_\_

(Insert completing mailing \_\_\_\_\_  
address, including country)

Typewritten Full Name  
of Fifth Joint

Inventor (if any) \_\_\_\_\_  
Given Name Middle Initial Family Name

\*\*Inventor's Signature \_\_\_\_\_  
\*Date of Signature \_\_\_\_\_  
Month Day Year

Residence \_\_\_\_\_  
City State or Province Country

Citizenship \_\_\_\_\_

Post Office Address \_\_\_\_\_

(Insert completing mailing \_\_\_\_\_  
address, including country)

Note to Inventor: Please sign name on line 2 exactly as it appears in line 1 and insert the actual date of signing on line 3.

This form may be executed only when attached to the first page of the Declaration and Power of Attorney of the application to which it pertains.